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(54) Title of the Invention:

METHOD OF ADHERING PROVIDING HEAT RESISTANCE AND ELECTRICAL CONDUCTIVITY

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(72) Inventor: Sadatoshi Nakazono

402 Iwato-shataku, 3-24 Iwato-cho, Minami-ku, Nagoya, Japan

(72) Inventor: Masayoshi Takahashi

209-103 Chuodai-danchi, 1856-10 Shirayama-cho, Kasugai, Japan

(71) Applicant: Tokai Konetsu Kogyo, Co., Ltd.

5-1, 3-chome, Uchikanda, Chiyoda-ku, Tokyo, Japan

SPECIFICATION

1. Title of the Invention

METHOD OF ADHERING PROVIDING HEAT RESISTANCE AND ELECTRICAL CONDUCTIVITY

2. Scope of the Claims

The method of adhering ceramic members to one another providing heat resistance and electrical conductivity comprising:

pulverizing powder of a silicide of at least one metal selected from the group consisting of Mo, W, Cr, Ta, Nb, V, Zr, or a mixture of powder of a metal of said group and metal silicon powder into powder, the diameter of which is 150 μm or less;

combining said powder and an organic binder to obtain a paste mixture and adhering temporarily ceramic members one another by using said paste mixture; and

welding said ceramic members by heating at from 1300°C to 1700°C.

3. Detailed Explanation of the Invention

The present invention relates to a method of adhering two ceramic members, especially a method of adhering one electrically conductive ceramic membrane with another.

Several methods of adhering in an electrically conductive manner one ceramic membrane with another have generally been used: (1) a metal thermal spraying method (metalicon); (2) a metalizing method, wherein a metal such as molybdenum is allowed to penetrate into ceramic in a gas such as hydrogen; (3) a non-electrolytic plating method; or (4) a method, wherein silver or platinum paste is used for bonding at the junction of ceramic members, or sliver or platinum is baked on a ceramic surface to form a metal surface on which through solder or silver solder one ceramic member is adhered to another. However, when the adhered parts formed by these methods are exposed to a high temperature in the air, they are melted and oxidized, and as a result, electrical conductivity disadvantageously decreases.

To improve this disadvantage, the applicant of the present invention previously

disclosed in Japanese Patent Application No. 53-3478 a method of bonding that comprises: combining metal silicon powder or silicon alloy powder with an organic binder to obtain a paste mixture; adhering temporarily both members by using said paste mixture; and welding said members by heating at from 1300°C to 1500°C.

The present invention provides a method of adhering providing heat resistance and electrical conductivity as well as stability that are superior not only to the conventional methods mentioned above but also to the method of bonding disclosed previously.

That is to say, the present invention is a method of adhering comprises: pulverizing powder of a silicide of at least one metal selected from the group consisting of Mo, W, Cr, Ta, Nb, V, Zr, or a mixture of powder of a metal of said group and metal silicon powder into powder, the diameter of which is 150 μm or less; combining said powder and an organic binder to obtain a paste mixture and adhering temporarily ceramic members one another by using said paste mixture; and welding said ceramic members by heating at from 1300°C to 1700°C in the air or under the atmosphere of argon or nitrogen or under vacuum. The method of the present invention provides further improved heat resistance and electrical conductivity as well as greater stability compared with the method of bonding previously disclosed. Furthermore, clay, SiO_2, etc. can be added as long as electrical conductivity is not affected for easier adhesion.

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Organic binders include poly(vinyl alcohol), methyl cellulose, starch, etc. and an aqueous solution of such is used. An amount of such organic binder to add is preferably from 0.1% to 2% based on the paste mixture by weight.

When an organic binder is added to powder of a silicide of a metal of said group or a mixture of powder of a metal of said group and metal silicon powder to mix well until a paste form is obtained, if the diameter of said powder material is 150 μm or more, smooth paste cannot be formed and temporary adhesion of the members becomes difficult, especially unsuitable for adhesion of tiny or complicated parts.

The resulting paste mixture can be applied to the junction parts to adhere of one ceramic member to another by such means as using a brush and screen printing. It is preferable that the members to be adhered are temporarily adhered one another by the adhesive ability of the paste mixture and then dried at around 120℃ to harden the paste mixture to some degree.

Finally, the temporarily adhered members to be adhered are heated at from 1300℃ to 1700℃ in the air or under the atmosphere of argon or nitrogen or under vacuum. By such heating, the organic binder is decomposed and said mixture is welded so that the two members are adhered.

The resulting junction of the ceramic members made by the present invention have not only superior heat resistance and electrical conductivity but also greater stability of the adhered section. Therefore, the method of adhering of the present invention is suitable, for example, to bond a heating element to a terminal in an igniter. Especially, it is suitable for adhering a micro heating element made of silicon carbide described in Japanese Patent Application No. 52-156074, "HEATING ELEMENT HAVING A WIRE OR BAND SHAPE," disclosed previously by the applicant of the present invention.

EXAMPLE 1

One part (by weight) of $MoSi_2$, the particle diameter of which was 140 μm or less, and 2 parts (by weight) of water were combined and grounded into fine powder in an alumina pot mill, which was dried to remove water. Then, 70 wt% of the resulting dried powder, 10 wt% of Kibushi Clay, and 20 wt% of a 5% of poly(vinyl alcohol) solution were combined and mixed in a mixer/grinder for one hour to obtain a paste mixture. Then,

said paste mixture 2 was applied to the silicon carbide terminal 1, the diameter of which was 3 m/m and the length of which was 40 m/m, penetrating and held through the insulator 4 as shown in Figure 4. Then, the separately prepared silicon carbide heating element 5 having a wire shape, the diameter of which was 0.5 m/m and the length of which was 10 m/m, was temporarily adhered to the paste mixture 2, and then dried at about 120°C, which was welded by heating at 1450°C in the air to obtain the resistor heating ignition element shown in Figure 2. This element heated to 1200°C under the load of 10 W, and after 1000 hours, a change in electrical resistance was within the range of ±8% and no abnormality such as a crack or peeling off was observed at the adhered section at all. Also, after the entirety of this element was left in the air at 1000°C inside a kiln for 1000 hours, a change in electrical resistance remained −5% or less and similarly no abnormality was observed at the adhered section.

EXAMPLE 2

Metal molybudenum powder, 50 wt%, the particle diameter of which was 43 μm or less, and 50 wt% of metal silicon powder, the particle diameter of which was 23 μm or less, were mixed. Such mixture, 80 wt%, and 20 wt% of a 2% methyl cellulose solution were mixed in a mixer/grinder for one hour to obtain a paste mixture. By using this mixture, an element was formed orderly from Figure 1 to Figure 2 in the same manner as EXAMPLE 1, and welded by heating in an argon gas at 1450°C to obtain an ignition element having great heat resistance and electrical conductivity.

EXAMPLE 3

WSi_2 that had been roughly ground in advance, the particle diameter of which was 220 μm or less, 80 wt%, and 20 wt% of a 2% poly(vinyl alcohol) solution are ground and mixed in a mixer/grinder for one hour to obtain a paste mixture, the particle diameter of which was 150μm or less. With such mixture, an element was formed orderly form Figure 1 to Figure 2 in the same manner as EXAMPLES 1 and 2, and heated under vacuum at 1600°C. This element heated by itself to 1200°C, and after 1000 hours, a change in electrical resistance was within the range of ±8.5%. Also, after the entirety of this element was left in the air at 1000°C inside a kiln for 1000 hours, a change in electrical resistance remained +3%.

Next, the element having a shape shown in Figure 2 formed by the method of

adhering in the present invention was compared with the element having the same shape formed by the method of bonding according to Japanese Patent Application No. 53-3478 and with the element having the same shape formed by the method of bonding with platinum, one of the conventional methods, and the comparison of characteristics is shown in Table below.

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Table

	According to the present invention	By the method of bonding according to Japanese Patent Application No. 53-3478	By the conventional method (the method of bonding with platinum)
A change in electrical resistance after heating by itself at 1200℃ for 1000 hours	±8.5%	±15%	+40%
A change in electrical resistance after being left in the air at 1000 ℃ for 1000 hours inside a kiln	±5%	±7%	∞
Stability at the adhered section (cracks, and peeling off, etc.)	99 to 100%	85%	75%

As shown above, the method of adhering in the present invention provides superior heat resistance and electric conductivity compared not only with the conventional methods but also with the previously disclosed method. In addition, the stability of the adhered section is close to 100%. In the past, disadvantages in heat resistance, electrical conductivity, and stability occurred on adhering one ceramic member to another. Any simple and inexpensive method of adhering has not been provided to satisfy such requirements, and demands for such method have been high in this field. The present invention, by which all of the problems related to the conventional methods are solved, can bring remarkable benefits in the various fields of high temperature technology.

Moreover, by using the paste mixture having great heat resistance, electrical conductivity, and stability according to the present invention, as an example of further advanced applications of the present invention, a printed electric circuit having greater heat resistance and electrical conductivity than the conventional ones can also be manufactured, wherein said paste mixture is screen printed on the surface of a ceramic insulating board and baked for adhesion.

4. Brief Description of the Drawings

Figures 1 and 2 are perspective views to explain each of EXAMPLES to which the method of the present invention is applied.

1 the silicon carbide terminal

2 the paste mixture

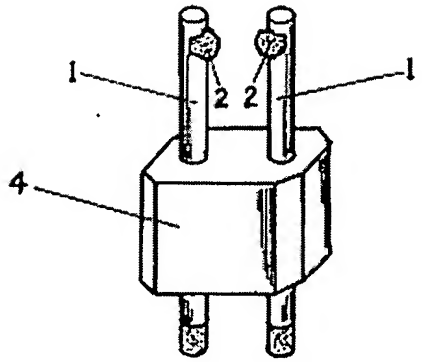
3 the silicon carbide heating element

4 the insulator

Applicant Tokai Konetsu Kogyo, Co., Ltd.

[Figure 1]

第 1 図



[Figure 2]

第 2 図